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PESTICIDE REMOVAL BY MEMBRANE PROCESSES

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ABSTRACT

Approximately 21 billion pounds of pesticides have been applied to United States farmlands since 1964. In agricultural regions, high pesticide concentrations occur in surface and groundwaters because of spring runoff or leaching. Because many of these compounds pose health risks, the United States Environmental Protection Agency (USEPA) has a mandate, under the Safe Drinking Water Act Amendments of 1986, to regulate several of these compounds. Others will be undergoing regulatory scrutiny because they are included in EPA's First Drinking Water Priority List, as well as the National Pesticide Survey. In an attempt to understand possible compliance problems, an investigation was conducted on river water containing Alachlor, Atrazine, Cyanazine, Linuron, Metolachlor, Metribuzin, and Simazine. The purpose of the study was to determine to what extent several treatment processes such as reverse osmosis were able to remove pesticides from the Sandusky River at the Tiffin, Ohio Water Treatment Plant. In addition pilot scale studies were conducted using several different types of polymeric membranes to remove pesticides from spiked groundwater.

INTRODUCTION

Between 1964 and 1986 approximately 21 billion pounds of pesticides were applied to United States (U.S.) farmlands (1). Of the 1.1 billion pounds of pesticides used in the U.S. in 1986, Agriculture used 75%, Industrial/Commercial/Government used 17%, and Home/Garden used 7% (1).

Before World War II, pesticides were almost exclusively inorganic compounds such as the arsenates, which accumulated in soils to high levels and eventually made fields unusable (2,3). After the war, new chemicals such as DDT, Aldrin, Endrin, and Dieldrin were introduced (2). Because of increasing public pressure associated with the effects of these compounds on wildlife and the environment, they were banned, thirty years later, in the early 1970s, in the U.S. and abroad (4,5). They were then replaced by less persistent, more biodegradable, but sometimes more toxic insecticides that included organophosphorous compounds and the carbamates (6). Interestingly, the pesticides most in use today, Atrazine and Alachlor herbicides, are not for control of insects but for control of weeds.

Many of these replacement pesticides were considered environmentally safer than their predecessors. Unfortunately, in addition to the direct effects of these compounds on the flora and fauna of the U.S., many found their way into the nation's water sources through agricultural runoff and leaching. Some of the compounds were removed from drinking water by conventional treatment because the pesticides adhered to particulates that were removed by settling and filtration. Others, assumed to be removed, were not. Some biodegraded in the environment. Others, assumed to biodegrade, seemed to persist and still remain in soils, muds, sediments, the biota, and foods, as well as in drinking water sources (rivers, lakes, streams, estuaries and wetlands). Some leached into groundwater, used by many rural communities, where they accumulated.

Surface Water and Groundwater Contamination

From the 1950s to the present, studies conducted in over 10 states and several countries outside the United States indicate possible drinking water contamination from both persistent organic pesticides, now banned, as well as the more modern pesticides. Although definite conclusions cannot be drawn from this information, Tables 1 and 2 illustrate the potential contamination of both surface water and groundwater sources by pesticides (3,6,7-23). Also brought forth in Table 1 (Column 6) is the general ineffectiveness of conventional treatment in removing many of these compounds from surface and groundwaters. In some agricultural states such as Iowa, pesticide contamination of surface and ground waters is well documented. A recent report documents the leaching of Atrazine, Alachlor, Cyanazine, and Metribuzin into the Karst-carbonate aquifiers of Northeast Iowa (24). The highest concentration of Atrazine occurred in mid-summer and early fall, corresponding to leaching after annual application of herbicides (24). Approximately 20% of the United States is underlain by limestone or dolomite, making these areas particularly sensitive to infiltration from pesticide-laden rivers or direct leaching (25). The U.S. has

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25-30 times as much ground water as surface water and half the population (about 95% of the rural population) depend on it for drinking water (26). River water infiltration of ground water is also of concern in many European countries such as Switzerland (27).

Regulations

Initially, under the Safe Drinking Water Act, National Interim Primary Drinking Water Regulations were set by the EPA that established Maximum Contaminant Levels (MCLs) for Endrin, Lindane, Methoxychlor, Toxaphene, 2,4-D, and 2,4,5-TP in 1975 (28). Because these pesticides were seldom, if ever, found in finished drinking water, and utilities were never found in noncompliance with the MCLs (29), pesticide contamination of drinking water was not well recognized. Attention has now shifted, however, to pesticides that present a far greater potential for being present in drinking water sources.

Simazine and Atrazine are part of the list of 83 contaminants required to be regulated under the Safe Drinking Water Act Amendments (SDWAA) of 1986 (30). Others, such as Metolachlor, Cyanazine, and Metribuzin, are on lists such as the EPA's First Drinking Water Priority List (FDWPL), as well as the National Pesticide Survey (Table 3)(30,31). Because of the large scale applications of pesticides in the U.S., many pesticide compounds have a strong potential to occur in public water systems. Perhaps, partly for this reason, the FDWPL will be reviewed and revised every 3 years. This may shorten the time between the first use of a pesticide or substituted pesticide and when it is found in drinking water (or when a regulation is considered).

Membrane Processes for Pesticides Control

One new technology that shows promise for control of pesticides is reverse osmosis. This membrane process has been used successfully for many years to desalt sea and brackish waters, to remove inorganics from drinking waters and to remove a wide variety of high-molecular-weight organics from industrial wastewaters. Figure 1 shows the theory of mass transport in reverse osmosis as well as the natural process of osmosis and evaporation/condensation. In evaporation/condensation pure water will vaporize and condense into the salt solution as water spontaneously moves from a region of higher vapor pressure to a region of lower vapor pressure until equilibrium is established. Similarly, one theory claims that in osmosis, water passes through a semipermeable membrane (from a region of high vapor pressure to a region of low vapor pressure) until equilibrium is reached. Reverse osmosis will remove organic contaminants from polluted water if pressure is applied to the contaminated side. This overcomes the osmotic pressure and clean water will pass through the membrane.

In the past, the membrane of choice was normally either cellulose acetate or polyamide. In the 1970s a new type of membrane appeared, the thin-film-composites. These membranes were produced from a variety of polymeric materials that were formed in-situ or coated onto the surface of an extremely thin, polysulfone support. Chian (32) conducted short term laboratory tests (small test cells) to evaluate cellulose acetate and cross-linked polyethylenimine membranes for removing Atrazine by high pressure RO (600psi). The NS-100 membrane, achieved 97.8% removal of Atrazine, and the cellulose acetate membrane achieved 84.0% removal of Atrazine.

Pilot scale tests conducted by the author indicate that some volatile organics and pesticides can be removed from distilled water and spiked ground waters by certain RO membranes (33). In another study, seven types of reverse osmosis membranes were comparatively tested on Suffolk County, New York groundwater (34). The units were tested for 5 to 24 weeks at pressures between 160-200 psi on groundwater contaminated with aldicarb, carbofuran, 1,2-dichloropropane, 1,2,3-trichloropropane and nitrate. The cellulose acetate and polyamide membrane removed some of the organics but not all. A spiral wound polyamide membrane showed the best removals (>85%) for pesticides and nitrates during the limited tests. The cost of RO to treat to below MCL's was estimated at between 1.95 and 5.00 dollars per thousand gallons for a 1 mgd. to .01 mgd. plant. Thus, this approach to treatment may not be economically unrealistic, especially when the combination of pesticides and nitrates are present (35).

BACKGROUND

In 1983, the Water Quality Laboratory of Heidelberg College conducted a pilot study on the occurrence of currently used pesticides at three municipal water supplies in Northwestern Ohio (35). These studies were undertaken in conjunction with tributary loading studies of sediments, nutrients, and pesticides that the laboratory conducts for the Great Lakes National Program Office of the U.S. EPA. The results of the 1983 program indicated that, for two water treatment plants lacking carbon filtration (Tiffin and Bowling Green), herbicides present in river water passed in undiminished concentrations through the treatment plants (36). At Fremont, Ohio where treatment includes the use of a granular activated carbon filter, herbicide concentrations were reduced considerably below river concentrations. These preliminary studies helped call attention to the human exposures to many currently used pesticides through drinking water.

In Seneca County, through which the Sandusky River flows (Figure 2), over one half million pounds of pesticides were applied to croplands in 1986 (41). Historically it has been assumed that these compounds were not persistent, yet in a controlled degradation test, Alachlor and Atrazine were shown to be persistent even after 68 weeks (1 yr. 4 months) (42). A report by Nicholson indicated chlorinated hydrocarbons were resistant to biological and natural chemical degradation, resulting in detection of residues several years after use in some areas of the U.S. (39). Table 4 shows the half-lives (in weeks) for hydrolysis and soil/field dissipation for Alachlor, Atrazine, and Simazine (40). The above factors should be used to calculate an environmental mass balance that would predict the pesticide contamination, in agricultural regions, of various water sources.

Tables 5 and 6 illustrate the peak concentrations of several currently used pesticides which were observed in northwestern Ohio from 1983 through 1988 (41). Alachlor, Atrazine, and Metolachlor, in several rivers and streams in northwestern Ohio, have the highest concentrations. Alachlor, atrazine and metolachlor corresponded, for example, to 26, 20 and 16.7 percent respectively, of the total Ohio herbicide usage in 1982 (41). This shows that insecticide and herbicide usage patterns may indicate potential surface water contamination. Some important factors influencing the timing of contamination are: rainfall, the dose applied and the frequency of pesticide application, the solubility/degradability of the pesticide, the soil characteristics, and the topography (42).

Results - 1984 Tiffin, Ohio Study

Tests were begun in 1984 in conjunction with The Ohio American Water Co. at the water treatment plants in Tiffin, Fremont, and Bowling Green, Ohio. The purpose of these tests was to determine the removal of alachlor, metolachlor, metribuzin, atrazine, simazine, cyanazine, and carbofuran from public water supplies by water treatment plants. This study focused on conventional and powdered activated carbon (PAC) treatment. Results showed the following (43):

* Water soluble pesticides were present in agricultural watersheds for up to six months (or longer) after application
* Pesticides were adsorbable on carbon but were not controlled by conventional treatment without carbon. The concentrations of pesticides in the raw river water samples were approximately the same as in the finished water samples.

EXPERIMENTAL

Objectives-1986, Tiffin, Ohio Study

In 1986, in order to collect more surface water occurrence data and additional treatment data on a wider variety of technologies (ozone oxidation, reverse osmosis, powdered activated carbon, aeration, etc.) another cooperative research effort was conducted with the Ohio American Water Co. at Tiffin, Ohio. This paper primarily addresses the reverse osmosis portion of this study as well as the in-house reverse osmosis research. The objectives were:

* Compare in-house, spiked, groundwater reverse osmosis data to field run surface water data.
* Using a finished-water side-stream, examine long-term, pilot-scale effectiveness of several types of commercially available reverse osmosis membranes on a field contaminated surface water over several months time.
* To verify mass balances and to determine the best polymeric membrane for removal of pesticides.

A thorough discussion of the conventional treatment results from concurrent studies, with and without PAC, can be found in several other reports (36,41,43,44,45). Ozone and aeration results can also be found in another report (46).

Reverse Osmosis

For the Tiffin study, the three pressure vessels, configured as shown in Figure 3, housed a thin-film-composite membrane, a cellulose acetate membrane, and a polyamide membrane. They were connected in parallel to the filtered river water supply inside the water treatment plant. Three membranes were chosen to compare the removal of the pesticides by different polymeric materials and their respective different module configurations. Operational pressures ranged from 150 to 200 psi, with influent flow rates ranging from 1.0 to 3 gal/min (4.2 to 11 Lpm) (Table 7). Filtered water was used because unfiltered water might contain particles that would quickly clog the membrane. The membranes were run from June 12, 1986 through July 30, 1986 during the 1986 spring runoff season. The influents and effluents from each unit were monitored frequently for total dissolved solids (TDS), pH, temperature, operating pressure, and flow rates. Influent, concentrate (reject), and permeate water from each unit were sampled daily for pesticide analysis, during the major runoff event which occurred in mid-June. This provided information for the mass balances.

Before and after the runoff event, the units were sampled once or twice per week. After the tests were completed, flushing tests were conducted. Distilled water was passed through each membrane for approximately one week. Permeate samples were collected daily, during the flushing period, in order to determine if membrane adsorption of pesticides had occurred.

Figure 4 is a schematic of the in-house pilot scale apparatus. Several membranes were connected in parallel and run for several days to several months. The interchangeable unit could accommodate different thin-film composite membranes. For example the FT-30 membrane was run first in the interchangeable unit for a length of time, flushed, and then removed. After these tests were analyzed, the DSI membrane was inserted into the interchangeable unit and run. Spiked groundwater was fed directly to each unit. Effluent water from all three units was recycled and used as feed water. See Table 7 for operating conditions.

RESULTS AND DISCUSSION

Table 8 compiles the reverse osmosis data collected from Tiffin and the in-house studies. Spiked groundwater was used for all in-house tests and conventional treated surface water (filtered water) used for the Tiffin runs. Spiked groundwater concentrations ranged from approximately 70 ug/L to 160 ug/L for Simazine, Atrazine, Linuron and Alachlor and 30 ug/L to 111 ug/L for Metolachlor. Tiffin filtered water concentrations ranged from none found to 3 ug/L for Simazine, Metribuzin, Cyanazine, Linuron and from none found to 15 ug/L for Atrazine, Alachlor and Metolachlor. Permeate concentration ranges are generally low in comparison to reject or influent concentration ranges for pesticides well removed by certain RO membranes. For example for the FT-30 in-house runs ,influent concentrations for Alachlor and Metolachlor were between 30 ug/L and 111 ug/L, the permeates were at none found and the concentrates ranged between 58 ug/L and 131 ug/L. The similarity between the influent ranges and the concentrate or reject ranges reflect the low permeate recovery of water flow. The same general trend can be seen for the FT-30 Tiffin permeate and concentrate ranges.

As with VOC removals in other studies (33), removals of pesticides varied greatly, according to the polymeric material of the membrane. Removals were calculated as:

$$\frac{\text{Influent Conc. } (\mu g/L) - \text{Permeate Conc. } (\mu g/L)}{\text{Influent Conc. } (\mu g/L)} \times 100 = \text{Percent Removal}$$

In some cases the final, steady state removals were used and in others, if more variation was evident, an average percent removal over time was used.

For the Tiffin study (Figure 5 and Figure 6) the cellulose acetate membrane removed less than 30% of the Linuron, Atrazine and Simazine, 40-50% of the Cyanazine, 59% of the Metribuzin and 70% of the Alachlor and 80% of the Metolachlor. The polyamide membrane showed removals consistently over 70% for the pesticides and 100% removal of Linuron. The thin-film-composite (TFC) membrane used at Tiffin, an FT-30, was superior to the cellulose acetate and polyamide membranes and removed almost 100% of Simazine, Metribuzin, Atrazine, Cyanazine, Linuron, Alachlor, and Metolachlor. The FT-30 membrane, manufactured by Filmtec Corporation, is a polyamide membrane that is cross-linked and contains carboxylate groups.

The in-house EPA studies using the cellulose acetate, polyamide, FT-30 membrane and another type of thin-film-composite, the DSI, also show about the same removals as the Tiffin removals. The DSI membrane is manufactured by Desalination Systems, Inc. and is a modified polyalkene on a polysulfone base on a backing of non-woven polyester. The chemical structure is proprietary. For most of the pesticides little difference is seen in removals of pesticides by the RO membranes from ground and filtered surface waters. For the In-house runs, TDS concentrations ranged for the spiked groundwater from 340 ppm to 795 ppm, pH from 7.4 to 8.2 and temperatures from 23 to 31 degree centigrade. For the Tiffin study, filtered surface water fed into the RO units had TDS concentrations that ranged from 202 ppm to 341 ppm, pH from 6.96 to 7.65 and temperatures from 21.2 to 28.9 degree centigrade. This indicates that water quality differences do not have a major impact. Additionally, variability in influent pesticide concentrations (Tiffin pesticide concentrations were much lower than the in-house concentrations) also did not appear to affect removals appreciably.

Differences in membranes are dramatically seen in removal of Linuron. Zero percent is removed by the cellulose acetate membrane, whereas 100% is removed by the polyamide membrane and the thin film composites. This clearly shows the importance of carefully choosing the proper polymer to remove certain individual or classes of compounds.

Mass balance were calculated as follows:

$$\frac{(\text{Influent Conc.(ug/L)})(\text{Influent Flow(L/min)}) - (\text{Mass OUT})}{(\text{Influent Conc.(ug/L)})(\text{Influent Flow(L/min)})} \times 100 = \% \text{ Mass Deviation}$$

where:

$$\text{Mass OUT} = [(\text{Perm. Conc.(ug/L)})(\text{Perm. Flow(L/min)}) + (\text{Rej. Conc.(ug/L)})(\text{Rej. Flow(L/min)})]$$

Mass balances were verified for all tests, and flushing tests indicated no major adsorption problems. Generally percent mass balance deviations for the in-house removals ranged between ± 20 and ± 30 percent (Figure 7). Much of this variability may be due to a combination of several factors such as the precision of pesticide analytics and the variability in reading gages for flow rates. In addition a slight adsorption/desorption phenomena may have taken place because of the varying influent concentrations for some compounds. The same applied to the Tiffin tests for some of the pesticides. Mass balances for some of the pesticides such as Metribuzin, Cyanazine, etc. exhibited greater variability due to the low influent concentrations. Based on these preliminary in-house and field tests, the newer membranes, in particular the thin-film composites do not appear to exhibit the major adsorption problems encountered with membranes tested years ago (32). Longer term tests of six months or more need to be conducted to verify the preliminary conclusions of this work. In addition longer term flushing tests should be conducted as well as an analysis of the membrane material itself to determine if pesticides are being adsorbed and desorbed. The main goals of these tests would be to verify mass balances, to identify membranes the least likely to exhibit removal variability, to show length of life of membranes and to verify that adsorption problems do not occur.

Percent recoveries (permeate flow rate/influent flow rate x 100) were between 9 and 23% for these small, one-pass pilot scale units. A trade off exists that affects eventual costs of the use of this technology. To keep plant size small, units are usually operated as multiple pass, in series to improve recoveries and reduce reject volume for disposal. Disposal of concentrated reject can be a problem. Alternately large units or units in parallel with low recoveries (as in this study) can be used in which case the reject concentration ranges are similar to the influent concentration ranges. If only 10-20% if the influent becomes permeate, that leaves 80-90% of the influent to be discharged as reject water that has approximately the same concentration as the influent. For these one pass systems, it may not therefore be unrealistic to recharge the reject water back into the source.

CONCLUSIONS

Seasonal applications of pesticides and fertilizers to agricultural lands have been identified as a significant source of non-point water contamination in the U.S. as well as abroad. If one pesticide is found, it is likely that there are others. Contaminant Level Goals have been proposed by the U.S. Environmental Protection Agency for several of these pesticides, and others are undergoing regulatory scrutiny. Tests were conducted in the Spring of 1986, at the Tiffin, Ohio Water Treatment Plant and in the author's laboratory, to identify treatment alternatives for communities. These results and results from other studies show that:

- Certain reverse osmosis membranes, especially the thin-film composites, have tremendous potential for treating dual contamination problems such as nitrates and pesticides. They can be used either on an intermittent basis, at a centralized location, or as a point-of-use/point-of-entry device. Conventional treatment consisting of clarification, filtration and chlorination was found to be ineffective for removal of the herbicides Alachlor, Atrazine, Cyanazine, Metribuzin, Metolachlor, Linuron and Simazine.
- Long term tests of six months or more need to be conducted to verify mass balances, to show length of life of membranes, to verify that serious adsorption problems do not occur and to choose the membranes that optimize removal and production of water without adsorbing pesticides.
- Membranes may be particularly useful in cases where individual wells in rural areas are contaminated from pesticides.
- Recovery of useable water for the one-pass units (in this study) tested was below 25% and the reject pesticide concentrations did not vary greatly from the influent concentrations. Therefore if a plant were to use multiple one-pass units, disposal of concentrate reject may not be a problem. In this case size may be a problem which ultimately affects cost, but not residuals disposal.
- In order to keep plant size small, a unit with multiple membranes in series and/or parallel could be used producing a very small volume of concentrated reject. In this case size would be small but residuals disposal would be a problem.

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Table 1 - Water Treatment Effectiveness and Pesticide Contamination of Ground and Surface Waters (1950-Present)

Date/Ref.	Compounds	Location	Water Type	Land Type, Erosion/Crop	Treatment	Removed by Treatment	Comments
1950/9	Toxaphene and other insecticides	Tenn. River, AL	S	cotton	no	no	
1955/10	Parathion and other insecticides	Tuscaloosa Co., Alabama	S	Poorly	no	no	Det. (ppb) in 15 streams from Toxaphene spraying after rains
1962/3	2,4-D and its ester derivatives	no	no	pilot study	no	no	Parathion present in water 4 months after application
1964/7	DDT, Lindane, Parathion, Dieldrin and 2,4,5 T Ester	no	S	pilot study	chemical coagulation, Cl_2 & $KMnO_4$ oxidation activated carbon conventional	ineffective effective effective	
1967/8	Atrazine	Pennsylvania	S	Silty clay loam corn	conventional ------ ozone - high doses	only DDT removed, not Lindane and Parathion ------ ozone oxidation but by-products toxicity	
1967/8	DDT, DDD, DDE, Toxaphene, Heptachlor Epoxide, Lindane, Dieldrin, BHC	California, east Texas, Sabine Basin, Miss., San Joaquin, Idaho, Ark	S	agriculture	no	no	
1969/9	Dieldrin, DDT, DDE	Missouri and Mississippi Rivers	S	no	no	no	Acid component suppressed agriculture runoff after rainfall
1971/11	Lindane	contaminated lake water	S	no	no	no	pesticides throughout aquatic environment due to major surface runoff
1969-72/12	Atrazine, endosulphan, DDT, DDE, HEOD, and 33 other pesticides	Canada	S	pilot study	ozone rapid sand filter activated carbon	no no no	30-40% of pesticide water samples had detectable levels of pesticides
1974/13	Atrazine, DDE, and Dieldrin	Rincon River and drain, Iowa	G/S	corn	conventional plants	ineffective	storm run-off from agricultural land and spills from mixing pesticides; all above pesticides in raw river water
							early major rains piled in high concentrations of pesticides; 2,4-D and Atrazine detected in raw water; 1/3 Rincon River composite pesticide data for Dieldrin from 1,000 to 5,000 ppb; drain water data pertinence of DDT.

G = Ground Water S = Surface Water no = not applicable or not available

Table 1 cont. - Water Treatment Effectiveness and Pesticide Contamination of Ground and Surface Waters (1950-Present)

Date/Ref.	Compounds	Location	Water Type	Land Type Source/Crop	Treatment	Removal by Treatment	Comments
1974/14	Lindane, heptachlor, Aldrin, p'-DDE, Dieldrin, pp'-DDD, pp'-DDT, and heptachlor epoxide	Chiomi River Tolentino, Italy	S	agriculture/widespread environmental contamination	conventional	some reduction, but pesticides never eliminated. Greatest decreases when turbidities highest	larger amounts of pesticides found in July and October.
1970's/15	Atrazine	Nebraska	G/S	sand/gravel mixed sand/gravel silty soils	na	na	sites in intensively irrigated counties adjacent to Platte River. Some shallow ground-water levels matched river concentrations.
1976/16	Heptachlor Epoxide and other pesticides	Lake St. Clair, Mt. Clemens, Michigan	S	Chlordane/Heptachlor applied to watershed	coagulation/settling GAC	source removal effective	samples collected during the period of runoff of season.
1976-77/17	Atrazine, triazines pesticides	Europe (Central)	S	corn, fruits, vineyards and fodder crops	na	na	June peak concentrations from runoff after spring application.
1970s/18	Alachlor, Atrazine, and Carbofuran and others	Kansas, Nebraska	G	tailwater pits and adjacent contaminated rivers	na	na	runoff water from pesticide treated fields is stored in tailwater pits for reuse as irrigation water, sometimes leaching into groundwater.
1970s/18	Atrazine	Iowa	G/S	agriculture	na	na	some well contamination associated with adjacent river that was contaminated by runoff.
1978/17	Atrazine	Rhode River, Maryland	S	local crops	na	na	
1980's/19	Lindane, Parathion, DBCP ethylene dibromide	Hawaii and/or California	G	agriculture	na	na	
1980's/19,20	Aldicarb Sulfoxide and Aldicarb Sulfone	central Wisconsin	G	Aldicarb applied to irrigated potatoes planted in sandy soils	na	na	peak nitrate concentration in shallow wells occurs in May, coincident with maximum runoff from fields.
1980's/15	Alachlor, Cyanazine, Atrazine and Metribuzin	N.E. Iowa S.E. Minn.	G	Leaching from soils into Incipient Karst and Karst Carbonate aquifers	na	na	
1980's/21	Aldicarb (sulfone, sulfoxide) Carbofuran, 1,2 DC Propane	Suffolk County Long Island, New York	G	pilot study/ field study	reverse osmosis and carbon	very effective	
1983/12	pesticide residues	Malaysia	S	fish culture wet rice cultivation	na	na	
1984/22	Atrazine, Cyanazine, Metolachlor, Alachlor, Carbofuran, Metribuzin, 2,4-D, Trifluralin, Butylate, Dicamba	Iowa	G/S	corn, soybean and other crops	conventional	ineffective for substantially removing pesticides from water samples	Samples collected after a rainfall event from 33 public water supplies. Concentration ranges from 0-26 ug/L.

G = Ground Water S = Surface Water na = not applicable or not available

2011

Table 2. Pesticides found in Groundwater in 23 States in the U.S.(23)

Pesticides	Type	State(s)	Range of Concentration, ppb
Alachlor	Herbicide	Maryl., Iowa, Neb., Penn.	0.1-10
Aldicarb (sulfoxide & sulfone)	Insecticide Nematicide	Ark., Ariz., Cal., Fl., Mass., Maine, N. Carol., New Jer., New York, Oreg., R.I., Texas, Vir., Wash., Wis.	1-50
Atrazine	Herbicide	Penn., Iowa, Neb., Wis., Maryl.	0.3-3
Bromacil	Herbicide	Florida	300
Carboluran	Insecticide Nematicide	New York, Wis., Maryl.	1-50
Cyanazine	Herbicide	Iowa, Penn.	0.1-1.0
Dibromochloropropane	Nematicide	Ariz., Cal., Hawaii, Maryl., S. Carol.	0.02-20
Dimethyl 2,3,5,6-tetrachloroterephthalate (and acid products)	Herbicide	New York	50-700
1,2-Dichloropropane	Nematicide	Cal., Maryl. New York, Wash.	1-50
Dinoseb	Herbicide	New York	1-5
Dyfonate	Insecticide	Iowa	0.1
Ethylene dibromide	Nematicide	Cal., Fl., Georgia, S Carol, Wash., Ariz., Mass., Conn.	0.05-20
Metolechlor	Herbicide	Iowa, Penn.	0.1-0.4
Metribuzin	Herbicide	Iowa	1.0-4.3
Oxamyl	Insecticide Nematicide	New York R. Island	5-65
Simazine	Herbicide	Cal., Penn., Maryl	0.2-3.0
1,2,3-Trichloropropane	Nematicide (impurity)	Cal., Hawaii	0.1-5.0

Table 3 - Proposed drinking water standards, regulatory lists and chemical class names for pesticides found in the Sandusky river water in Tiffin, Ohio.

Pesticide	Chemical Class	Proposed MCL ug/L *	1986 Safe Drinking Water Act [a]	First Drinking Water Priority List [b]	National Pesticide Survey [c]
Alachlor	acetamide	2			
Metolachlor	acetamide			X	X
Atrazine	triazine	2	X		X
Cyanazine	triazine			X	X
Simazine	triazine		X		X
Metribuzin	triazine			X	X
Linuron	urea derivative				X

a) Part of list of 83 contaminants to be regulated under the Safe Drinking Water Act of 1986 (30).
b) Part of list of 53 compounds on EPA's First Drinking Water Priority List (which indicates compounds may have adverse health effects, that they are known or anticipated to occur in public water systems and that may be regulated under the Safe Drinking Water Act of 1986) (30).
c) Part of list of 127 pesticides included in the National Groundwater Pesticide Survey (which will provide additional occurence information for pesticides over the next 2 years)(31).

Table 4. Persistence of Alachlor, Atrazine, and Simazine in water and soil (40)

Pesticide	Solubility (ppm)	Half-life(water) Hydrolysis (weeks)	Half-life(wks) Soil field dissipation
Alachlor	240	NA	1-10
Atrazine	33	10-106	<4-57
Simazine	3.5	8-30	<4-16

Table 5. Peak Observed Concentrations of Major Herbicides at the Monitoring Station During the Interval from April 1983 to October 1987. All Concentrations are given in ug/L. The First Stations are listed in the Order of Decreasing Watershed Size.

River or Creek	Watershed Size Km^2	# of Samples	Alachlor	Metolachlor	Atrazine	Cyanazine	Metribuzin	Linuron
Maumee	16395	340	25.85	28.66	15.61	13.27	8.61	8.79
Sandusky	3240	375	35.88	37.34	32.81	26.49	13.82	8.27
Honey Cr.	386	534	77.28	125.98	67.85	21.02	13.31	18.67
Rock Cr.	88	477	32.96	127.52	57.70	33.03	23.81	13.65
Lost Cr.	11.3	410	91.47	83.74	281.41	30.17	37.54	16.19
Cuyahoga	1831	94	1.64	7.09	3.80	1.81	1.57	6.08
River Raisin	2699	134	10.59	7.77	16.62	5.00	3.67	2.32

Table 6. Annual Variations in Peak Observed Concentrations of Atrazine, Alachlor and Metolachlor During the Pesticide Runoff Season for the Maumee and Sandusky Rivers and for Honey Creek.

	Alachlor, ug/L			Metolachlor, ug/L			Atrazine, ug/L		
Year	Maumee	Sandusky	Honey Cr.	Maumee	Sandusky	Honey Cr.	Maumee	Sandusky	Honey Cr
1983	10.54	6.94	12.49	9.25	21.97	30.82	7.22	10.63	23.30
1984	17.64	8.75	22.01	13.73	19.45	35.41	13.62	10.15	37.46
1985	5.63	26.52	27.06	8.52	42.36	35.36	9.00	28.20	29.33
1986	8.70	33.52	72.17	31.57	39.00	138.76	13.16	32.38	66.96
1987	7.66	13.18	74.14	13.71	26.12	30.86	13.05	21.64	46.20
1988	0.76	0.40	7.07	1.72	0.73	7.96	3.00	2.19	8.84

Table 7. Operating Parameters for Reverse Osmosis Units Used at Tiffin, Ohio Water Treatment Plant and EPA In-House Pilot Scale Tests

TIFFIN	EPA	MEMBRANE (CONFIGURATION)	DIAMETER		SURFACE AREA		OPERATING PRESSURE[1]		INFLUENT[3]		FLOW RATE[2] REJECT		PERMEATE		PERCENT RECOVERY[4]	PERMEATE FLUX	
			in	(cm)	ft^2	(m^2)	psi	(kPa)	gal/min	(L/min)	gal/min	(L/min)	gal/min	(L/min)		$gal/ft^2 \cdot day$	$(L/m^2 \cdot day)$
X	X	CELLULOSE ACETATE (Spiral Wound) 40 inch length	2	(5.1)	19	(1.8)	200	(1380)	1.1	(4.2)	1.0	(3.8)	0.1	(0.4)	9.1	7.58	(320)
X	X	POLYAMIDE (Hollow Fiber) 17 inch length	4	(10.2)	3000	(279)	150	(1035)	1.3	(4.9)	1.0	(3.8)	0.3	(1.1)	23.1	0.14	(5.7)
X	X	THIN FILM COMPOSITE (interchangeable unit) FT-30 (Spiral Wound) 40 inch length	2.5	(6.4)	25	(2.3)	200	(1380)	2.9	(11.0)	2.5	(9.5)	0.4	(1.5)	13.8	23.04	(939)
	X	THIN FILM COMPOSITE (interchangeable unit) DSI (Spiral Wound) 40 inch length	2.5	(6.4)	20	(1.86)	200	(1380)	2.62	(10.07)	2.5	(9.5)	0.32	(1.2)	11.2	23.04	(939)

[1] Operating pressures within ± 5%

[2] Flow rates within ± 10%

[3] INFLUENT = (REJECT) + (PERMEATE)

[4] PERCENT RECOVERY = (PERMEATE FLOW RATE)/(INFLUENT FLOW RATE) X 100%

Table 8. Influent, Permeate, and Concentration Ranges for In-House Groundwater and Tiffin, Ohio Surface Water Reverse Osmosis Tests.

Cellulose Acetate Membrane

Spiked Ground Water/In House

Pesticide	Influent Range ug/L	Permeate Range ug/L	Reject Range ug/L	% Removal	SS[a]	AVG[b]	# Samples[c]	Mass Balance Range
Simazine	85.1-117.2	30.4-64.9	84.5-126.4	31	X		21	9.3-11.2
Metribuzin	-	-	-	-				-
Atrazine	88.5-161.3	84.3-84.9	88.5-141.2	36-50		*		-4.4-31.6
Cyanazine	-	-	-	-				-
Linuron	74.7-108.6	84.8-87.9	76.5-92.3	0	X		6	-8.9-2.3
Alachlor	-	-	-	-				-
Metolachlor	-	-	-	-				-

Filtered Surface Water/Tiffin

Influent Range ug/L	Permeate Range ug/L	Reject Range ug/L	% Removal	SS[a]	AVG[b]	# Samples[c]	Mass Balance Range
0.11-0.616	0.09-0.94	0.13-0.94	23		X	14	33.56-16.22
0.9-2.727	0.0-0.73	0.04-3.21	50*		*		-31.12-31.29
2.48-11.752	1.61-8.85	2.9-13.48	29		X	15	-18.76-12.27
0.9-2.534	0.9-1.50	0.0-2.87	40-50		X	10	-27.85-4.31
0.9-1.179	0.9-1.08	0.9-1.35	0.0		X	8	* *
0.783-8.437	0.29-2.22	0.87-7.75	70		X	15	-20.97-15.22
2.731-14.808	0.0-4.1	2.27-20.32	60		X	15	-25.9-12.57

Polyamide Membrane

Spiked Ground Water/In House

Pesticide	Influent Range ug/L	Permeate Range ug/L	Reject Range ug/L	% Removal	SS[a]	AVG[b]	# Samples[c]	Mass Balance Range
Simazine	85.1-117.2	14.3-70.4	88.5-140	66	X		10	-10.8-7.7
Metribuzin	-	-	-	-				-
Atrazine	88.5-161.3	31.1-48.9	88.9-163.9	66		X	8	-19.5-23.0
Cyanazine	-	-	-	-				-
Linuron	74.7-108.6	31.8-53.3	82.5-118.4	57	X		6	-18.9-8.3
Alachlor	-	-	-	-				-
Metolachlor	-	-	-	-				-

Filtered Surface Water/Tiffin

Influent Range ug/L	Permeate Range ug/L	Reject Range ug/L	% Removal	SS[a]	AVG[b]	# Samples[c]	Mass Balance Range
0.11-0.616	0.0-0.1	0.15-1.04	72	X		13	-34.16-15.15
0.9-2.727	0.0-0.74	0.0-3.54	70	X		13	* *
2.48-11.752	0.3-4.11	3.42-16.53	70	X		13	-19.46-13.25
0.9-2.534	0.9-1.02	0.9-3.30	80	X		11	* *
0.9-1.179	0.9-1.94	0.9-1.37	100	X		10	* *
0.783-8.437	0.9-4.53	0.86-10.87	77	X		13	* *
2.731-14.808	0.37-3.27	3.9-23.4	70	X		13	-25.46-16.47

TABLE 8 CONTINUED

Thin Film Composite FT-30 Membrane

Spiked Ground Water/In House

Pesticide	Influent Range ug/L	Permeate Range ug/L	Reject Range ug/L	% Removal	SS[a]	AVG[b]	# Samples[c]	Mass Balance Range
Simazine	-	-	-	-				-
Metribuzin	-	-	-	-				-
Atrazine	-	-	-	-				-
Cyanazine	-	-	-	-				-
Linuron	-	-	-	-				-
Alachlor	73 4-108 0	0 0-0 0	81-125	100	X		22	-15 3-16 2
Metolachlor	30 9-111 0	0 0-0 0	56-131	100	X		22	-18 3-13 6

Filtered Surface Water/Tifts

Influent Range ug/L	Permeate Range ug/L	Reject Range ug/L	% Removal	SS[a]	AVG[b]	# Samples[c]	Mass Balance Range
0 11-0 816	0 0-0 3	0 0-1 36	100	X		18	+ +
0 0-2 727	0 0-0 05	0 0-3 54	100	X		18	+ +
2 48-11.752	0 0-0 58	2 68-14 81	98	X		18	-17 12 15 48
0 0-2 534	0 0-0 15	0 0-3 41	100	X		14	+ +
0 0-1 179	0 0-0 08	0 0-1 48	100	X		11	+ +
0 783-6 437	0 0-0 25	0 98 0 1	100	X		18	-23 49-17 66
2 731-14 806	0 0 1 0	3 07-17 42	100	X		18	-21 38-14 12

Thin Film Composite DS5 Membrane

Spiked Ground Water/In-House

Pesticide	Influent Range ug/L	Permeate Range ug/L	Reject Range ug/L	% Removal	SS[a]	AVG[b]	# Samples[c]	Mass Balance Range
Simazine	85 1-117 2	0 0-6 2	93 7-131 8	99	X		20	-3 6 0 8
Metribuzin	-	-	-	-				-
Atrazine	98 5-161 3	0 5-40	105 5-145 2	60-100*		*		-11 6 30 0
Cyanazine	-	-	-	-				-
Linuron	74 7-105 6	0 6-0 8	82-116	99	X		8	-12 4-9 5
Alachlor	-	-	-	-				-
Metolachlor	-	-	-	-				-

* Removal declined over time or fluctuated

- Tests not conducted

+ + Mass balance variations due to low influent concentrations

a = Final, Steady State percent Removal

b = Average percent removal (at Steady State)

c = Number of samples used to calculate average percent removal or number of samples over which steady state conditions were maintained .

EVAPORATION / CONDENSATION

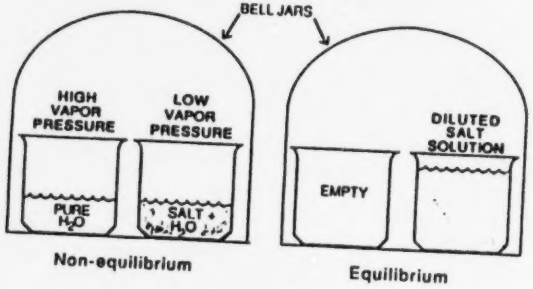
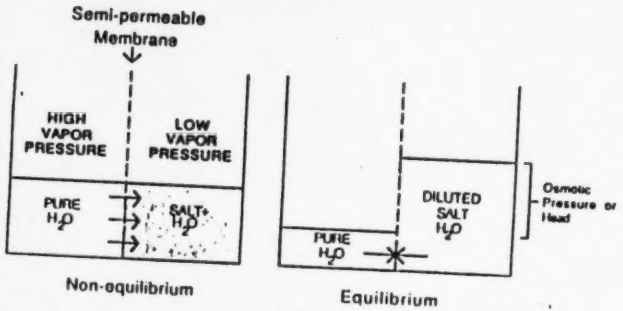


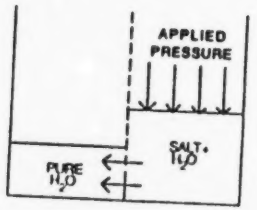
Figure 1. Pure water will vaporize and condense into the salt solution as water spontaneously moves from a region of higher vapor pressure to a region of lower vapor pressure until equilibrium is established. Similarly, one theory claims that in osmosis water passes through a semipermeable membrane until equilibrium is reached. Reverse Osmosis will remove organic and inorganic contaminants from polluted water if pressure applied to the contaminated side.

OSMOSIS



Tendency for water to pass through membrane = pressure exerted by column of liquid.
Applied pressure = Osmotic pressure

REVERSE OSMOSIS



Pure water is removed from salt solution or contaminated water when Applied Pressure > Osmotic Pressure.

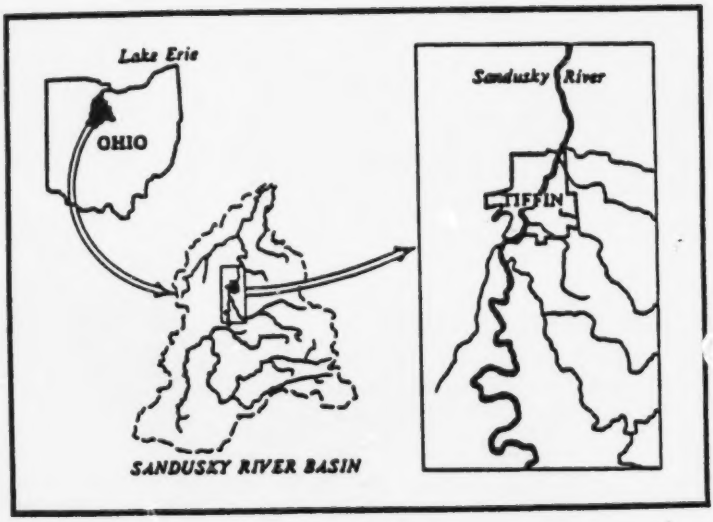


Figure 2. Location of the Sandusky River Basin, the Sandusky River, and Tiffin Ohio.

Figure 3. Flow diagram of the Tiffin, Ohio Water Treatment Plant Showing the sampling points and flow schemes for the pilot-scale reverse osmosis units.

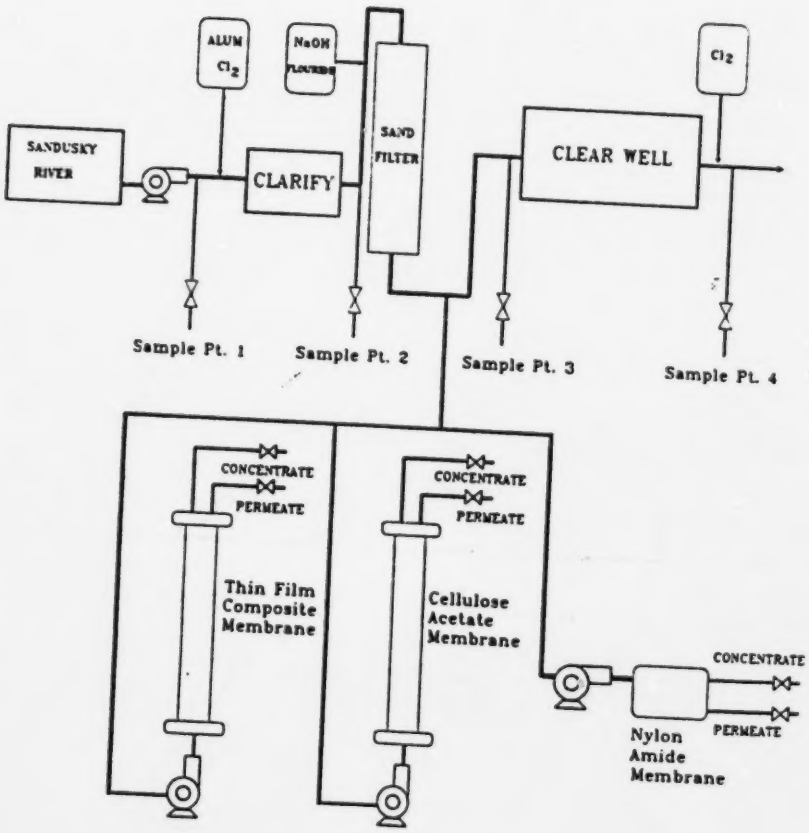


Figure 4. Flow Diagram of In-House Pilot-Scale Setup Showing each RO unit's Sampling Points

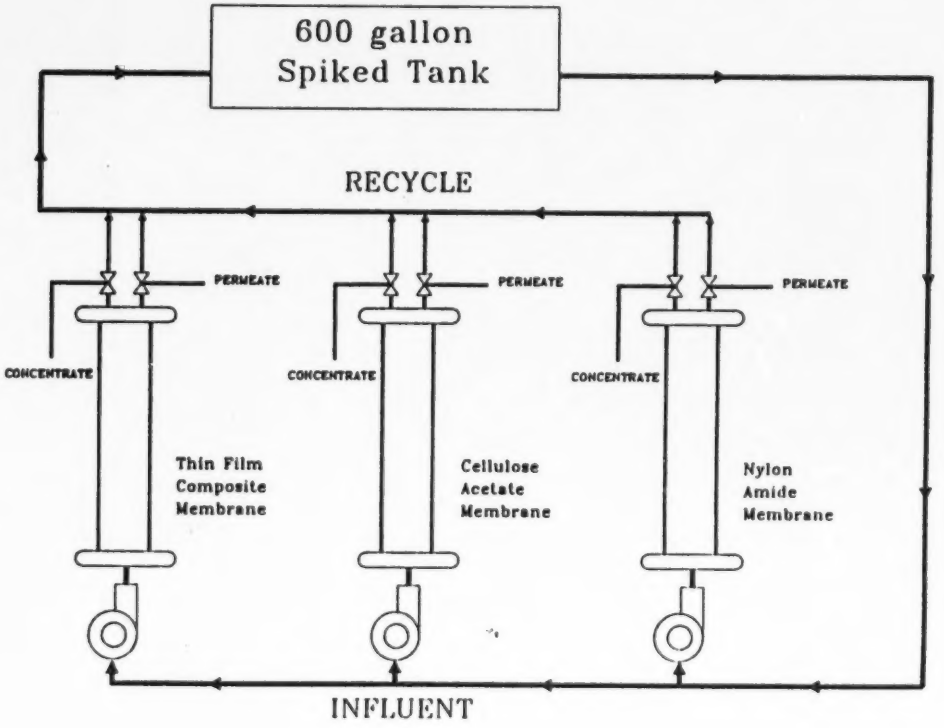
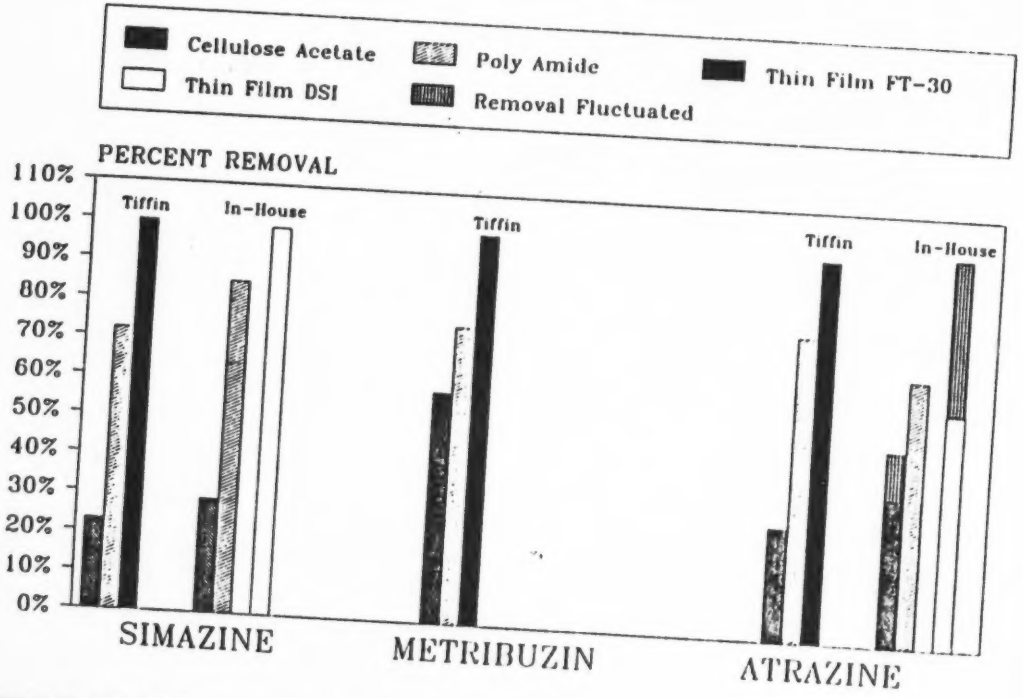


Figure 5. Percent Removal of Simazine, Metribuzin and Atrazine by a Variety of Reverse Osmosis Membranes.



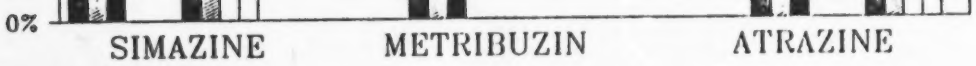


Figure 6. Percent Removal of Cyanazine, Linuron, Alachlor and Metolachlor by a variety of Reverse Osmosis Membranes.

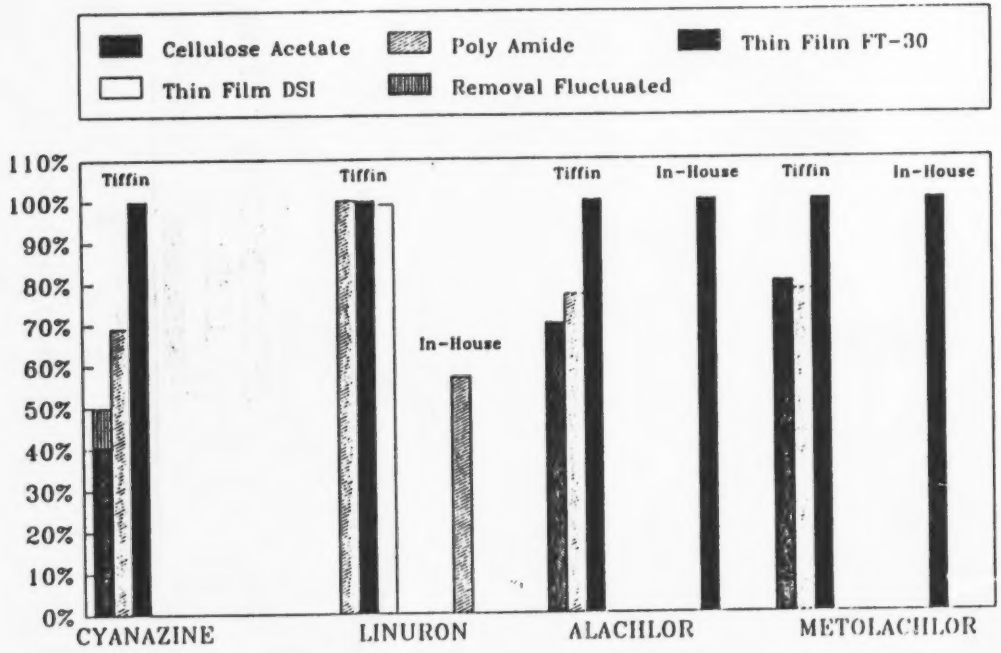


Figure 7. Mass Percent Deviation for In-House and Tiffin Reverse Osmosis Tests.